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Phase morphology, crystallisation behaviour and mechanical properties of isotactic polypropylene/high density polyethylene blends

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Abstract

The phase morphology, crystallisation behaviour and mechanical properties of isotactic polypropylene (iPP)/high density polyethylene (HDPE) blends were investigated. It was found that the properties are intimately related to each other. The morphology of the blends showed a two phase structure in which the minor phase was dispersed as domains in the major continuous matrix phase. The domain size of the dispersed phase increased with increasing concentration of that phase due to coalescence. It was also found that the domain size of the dispersed phase depends on the viscosity difference between the two phases. For a given HDPE/iPP blend, where HDPE is the matrix and iPP is the dispersed phase, the iPP domains were smaller than HDPE domains of the corresponding iPP/HDPE blend where iPP is the matrix and HDPE is the dispersed phase. A co-continuous morphology was observed at 50/50 PP/HDPE composition. Crystallinity studies revealed that blending has not much effect on the crystalline melting point of polypropylene and high density polyethylene. The crystallisation enthalpy and heat of fusion values of HDPE and PP in the blend were decreased as the amount of the other component increased. The variation in percent crystallinity of HDPE and PP in the blend was found to depend on the morphology of the blend. All the mechanical properties except Young's modulus and hardness showed negative deviation from the additivity line. This is due to the incompatibility of these blends.

Keywords: iPP; HDPE; Morphology; Crystallisation; Mechanical property

1. Introduction

Plastic waste has become one of the main issues of environmental concerns. Major polymeric components of plastic waste are polyethylene and polypropylene. Separation of plastic waste into individual polymers is costly and complete sorting is some times impossible. Plastic waste can be recycled easily by converting it into the form of polymer blends. Polymer waste recovery is providing greatly increased motivation for improving the properties of polymer blends. The combination of polyethylene and polypropylene is particularly important because it is very difficult to separate these polymers from each other in waste recovery operations. The

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difficulty arises due to the structural similarity of these polymers. The blends of polyethylene and polypropylene are commercially very important because of their high impact strength and low temperature toughness, furthermore, these polyolefins are the major constituents of plastic wastes and scraps and their recycling as blends is of considerable value. Addition of polyethylene into polypropylene increases the impact strength of polypropylene and addition of polypropylene into polyethylene improves the environmental stress crack resistance of polyethylene. Hence these blends are technologically very important.

The physical, optical and mechanical properties of polymer blends are affected to various degrees by the phase morphology. Mechanical properties such as tensile strength, tensile elongation and impact strength for a particular polymer blend vary with the morphology and to a lesser extent with the crystallinity. The morphology [1–5], mechanical properties [6–18] and crystallisation behaviour [19–21] of PE/PP blends were reported by several researchers. Gohil [12] has found a synergism in mechanical properties through epitaxial growth in PE/PP blends. Spardaro and Rizzo [14] have observed that mixing parameters have significant effects on the mechanical properties and suggested that it is very important to find optimum mixing procedures for each kind of blends. Lee et al. [17] found that mechanical properties of PE/PP blends are closely related to the morphology. Montes et al. [22] investigated the phase separation in blends of PE and PP and observed the phase separation for a wide range of compositions for temperatures in the range 170–200 °C. However the phase separated region became narrow slightly as the melt temperature is reduced.

It is important to note that even though morphology, crystallisation behaviour and the mechanical properties of these blends have been reported several times in the literature, there is no systematic study to correlate the morphology and crystallisation behaviour with the mechanical properties. Even though in the present investigation, virgin polymers have been utilized, our prime objective is to develop the best compatibilisation strategy to attain maximum improvement in ultimate properties and then to extend the study to commingled polyolefin waste so that it is possible to obtain recycled blends with desirable end use properties acceptable for some commercial applications. In order to attain this goal, one has to know the critical factors on which these properties rely and thus develop a thorough understanding on the phase morphology and crystallisation behaviour of the blends which can play decisive role in determining these properties. Therefore the present contribution is devoted to correlate the observed mechanical properties of PP/HDPE blends with their phase morphology and crystallisation behaviour. Scanning electron microscopy (SEM) was used for investigating the phase morphology of the blends and differential scanning calorimetry (DSC) for the in-depth interpretation of the crystallisation behaviour of the blends.

2. Experimental

2.1. Materials and blend preparation

Isotactic polypropylene (Koylene 3060) having an MFI of 3 g per 10 min and density of 900 kg m^{-3} was kindly supplied by Indian Petro Chemicals Limited, Baroda, India. High density polyethylene (HDPE-Relene, M60 200) having an MFI 20 g per 10 min and density 960 kg m^{-3} was supplied by Reliance Industries, India. Blends of different compositions of iPP and HDPE (H_{10}, H_{20}, H_{30}, H_{40}, H_{50}, H_{60}, H_{70}, H_{80} and H_{90}, where H denotes HDPE and subscript denotes the percentage composition by weight of HDPE) were prepared by melt mixing of PP with HDPE in a Brabender Plasticorder at 170 °C. The rotor speed was 60 rev min^{-1} and mixing time was 5 min. The melt mixed samples were then compression moulded at 185 °C.

2.2. Mechanical property testing

Tensile specimens were punched out from the compression moulded sheets. Tensile tests were performed in accordance with ASTM D412-80 test method using dumb-bell shaped test pieces at a cross-head speed of 50 mm/min using a Zwick Universal Testing Machine (Model 1474). The tear strength was also performed on Zwick UTM according to ASTM D624-81 using unnicked 90° angle test pieces at a cross-head speed of 50 mm/min. The notched Izod impact test was carried out in accordance with ASTM D 256 using an impact tester (Atsfaar, IMPATS-15, Italy). Specimens of 2 mm thickness and 3 mm ligament length were used. Hardness was measured according to ASTM test method using a Shore D Durometer. For hardness measurement, sufficient number of sheets were placed one over another to get a thickness of at least 6 mm, taking care that the surface of the sheets was flat. All the mechanical tests were performed at room temperature.

2.3. Phase morphology measurements

The morphology of the blends was analysed using a Jeol scanning electron microscope (Model-JSM-IC 848, 20 kV accelerating voltage). The samples for the morphology measurements were prepared by cryogenically fracturing the samples in liquid nitrogen. The size of the dispersed phase was analysed by image analysis technique. About 400 particles were considered for the diameter measurements. The number, weight, surface and volume average diameters were determined using the following equations:

The number average diameter

$$\overline{D}_{\mathrm{n}} = \frac{\sum N_i D_i}{\sum N_i} \tag{1}$$

The weight average diameter,

$$\overline{D}_{\mathrm{w}} = \frac{\sum N_i D_i^2}{\sum N_i D_i} \tag{2}$$

The surface average diameter,

$$\overline{D}_{\mathrm{z}} = \sqrt{\frac{\sum N_i D_i^2}{\sum N_i}} \tag{3}$$

The volume average diameter,

$$\overline{D}_{\mathrm{v}} = \frac{\sum N_i D_i^4}{\sum N_i D_i^3} \tag{4}$$

2.4. *Differential scanning calorimetry*

The melting and crystallisation behaviour of HDPE/PP blends was evaluated using a Seiko DSC thermal analyser (Model SSC 5200). The samples were scanned at a heating rate of 10 °C min^{-1} in nitrogen atmosphere. The DSC thermal history of the samples is represented in Fig. 1.

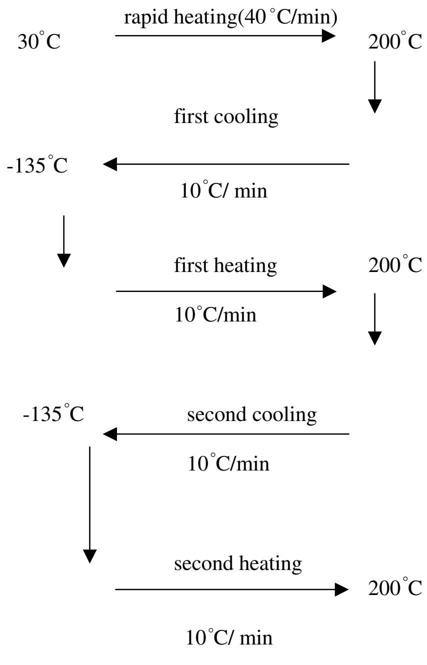

Fig. 1. DSC thermal history of the samples.

3. Results and discussion

3.1. *Phase morphology studies of PP/HDPE blends*

Phase morphology of heterogeneous blends is the major determinant of final properties. A clear picture of phase morphology of polymer blends was derived from several studies during seventies and early eighties. The important studies include those of Callen et al. [23], Nielsen [24], Danesi and Porter [25], Paul and Barlow [26], Han [27], Roland and Bohm [28] and Elmendrop and Maalche [29]. These studies predicted that the final morphology is determined by blend ratio, melt viscosity, processing parameters, presence of other ingredients, etc. Thereafter, several investigations were undertaken to develop empirical rules for the prediction of the phase structure in immiscible blends from the knowledge of their composition, component properties and the flow field in a mixing or processing device [30–35]. However no reliable theory or empirical rule explaining the dependence of phase structure on these parameters of a system has been derived so far.

The scanning electron micrographs in Fig. 2(a–g) show the phase morphologies of PP/HDPE blends. As mentioned earlier, these blends are immiscible and incompatible. For the blend containing 90% PP (Fig. 2a), HDPE phase is dispersed in PP matrix as spherical domains and the domain diameters are less than 1 μm. The domain diameters (number average, weight average, volume average and surface average) are plotted against blend ratio and are shown in the Fig. 3. As the percentage of HDPE increases, the size of the dispersed HDPE domains increases. This is due to the coalescence phenomenon. Coalescence, the recombination of particles, is known to take place during the mixing process and must arise from the forced collisions of dispersed particles [36]. If we assume the collisions in the melt are similar to the collisions in gases as predicted by kinetic theory of gases (remember that in gases, molecules are very free to move but in viscous liquids, the movements are so restricted that only neighbouring domains have a chance to recombine), the rate of collisions (here collision means only that particular state at which two domains are in contact with each other) is directly proportional to the square of the concentration of the domains. As the concentration of the dispersed phase increases, the concentration of the domains also increases and consequently the rate of collisions of these dispersed particles increases. The net effect is a marked increase in the domain size due to coalescence. This is why the particle size increases as the weight percentage of HDPE in PP rich blend increases. For the blend containing 90% PP, due to the low concentration of dispersed HDPE phase, the rate of coalescence is comparatively low and consequently the size of dispersed HDPE domains is relatively small.

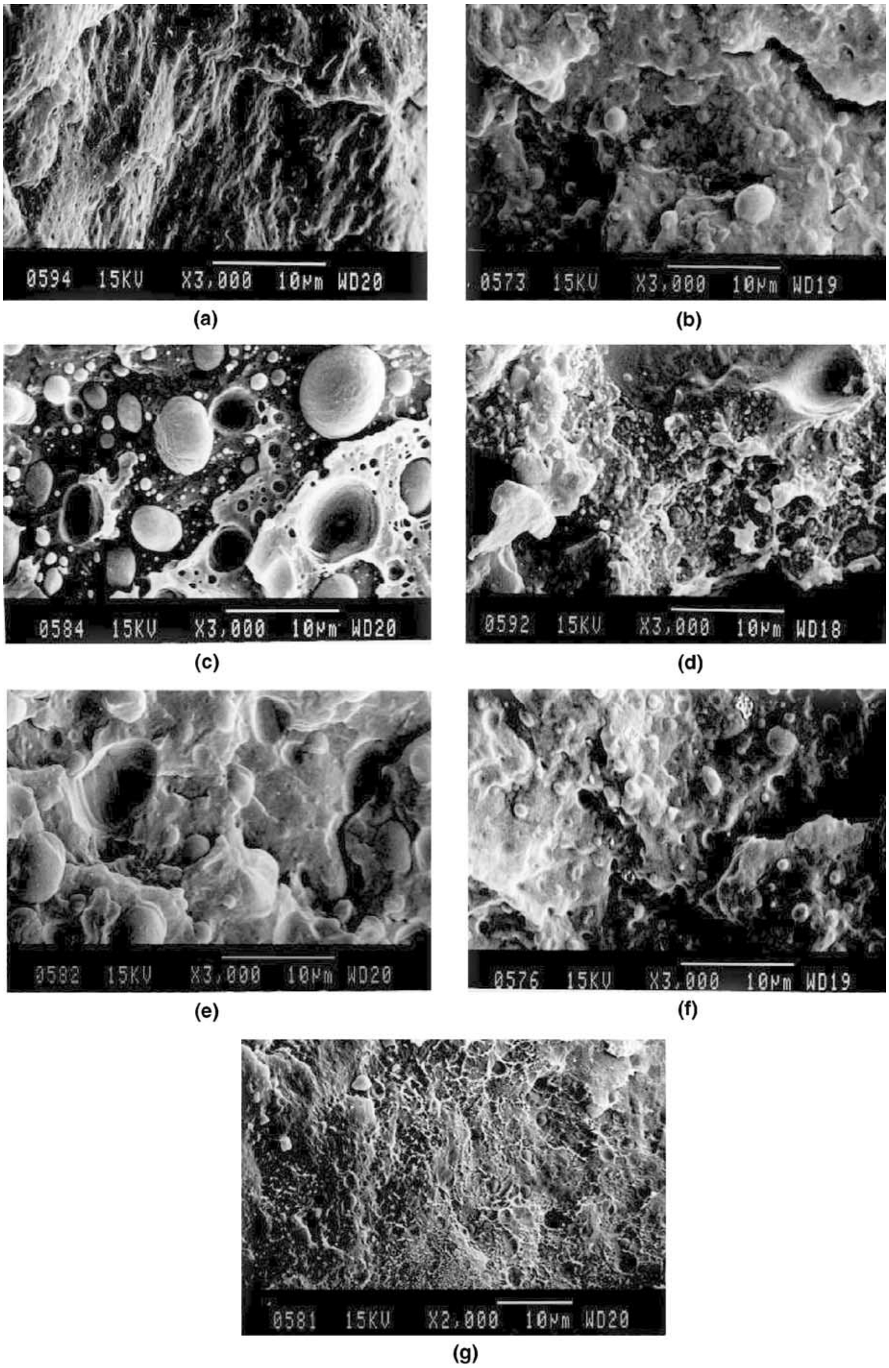

Fig. 2. Scanning electron micrographs showing the morphology of (a) H_{10}, (b) H_{20}, (c) H_{30}, (d) H_{50}, (e) H_{70}, (f) H_{80} and (g) H_{90}.

Thomas and Groeninckx [37] explained the various steps in coalescence. The first step is the diffusion of dispersed domains through the matrix, second one is the collision between the dispersed domains and the final step is the coalescence itself. Since the diffusion and coalescence phenomena are fast and the collision is a comparatively slower process, the collision between the dispersed domains may be taken as the rate-determining step. From this, it is evident that coalescence can be decreased by controlling the collisions (contact between the domains) between the dispersed domains. Based on the fact that, blending involves the continuous break-

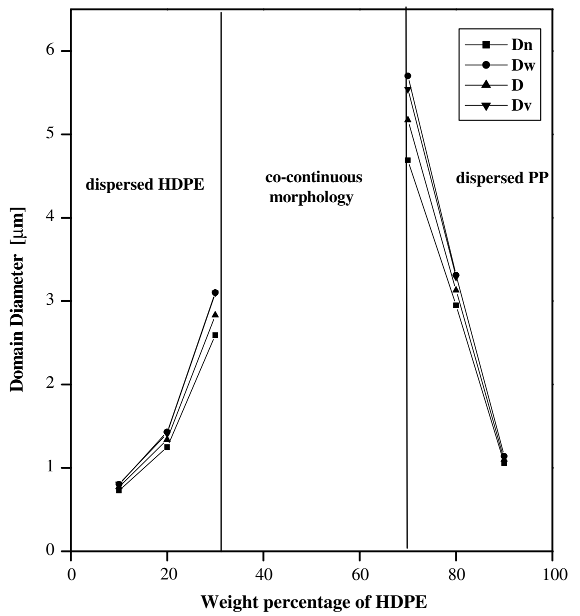

Fig. 3. Variation in average particle size as a function of blend composition.

down and coalescence of the dispersed particles, Tokita [38] derived an expression for analysing the particle size of a dispersed phase in polymer blends. The equilibrium particle size is achieved when the rate of breakdown and coalescence are balanced.

The rate constant of breaking drop is

$$K_1 = \frac{\eta\dot{\gamma}^2}{E_{\mathrm{dk}} + 3v/R} \tag{5}$$

The rate constant for coalescence was calculated as

$$K_2 = \frac{4p\phi_{\mathrm{d}}\dot{\gamma}}{\pi} \tag{6}$$

At equilibrium, when breakdown and coalescence are balanced, $K_1 = K_2 = 0$. The equilibrium particle size R^* may be expressed as

$$R^* = \frac{12pv\phi_{\mathrm{d}}}{\pi\sigma}\left(1 - \frac{4p\phi_{\mathrm{d}}E_{\mathrm{dk}}}{\pi\sigma}\right) \tag{7}$$

where R^* = radius of particles, σ = shear stress, v = interfacial tension, γ = shear rate, ϕ_{d} = volume fraction of the dispersed phase, E_{dk} = bulk breaking energy and p is the probability that a collision will result in a coalescence.

From this equation it is clear that the particle size will be smaller when the volume fraction of the dispersed phase is smaller. Again, as the concentration of the dispersed phase decreases, the probability that a collision will result in coalescence becomes minimum. As the particle size increases the probability also increases. At one particular state it reaches a maximum. This critical state where there is maximum probability for a collision to result in coalescence may be same for many systems provided there is no marked difference in the viscosity between the two component polymers. This is because at this state, the particles are very close to each other. Hence the first step (diffusion of particles through the matrix) is not very significant. After this point the blends are said to possess a co-continuous morphology. Thus we can predict the critical state, where there is minimum coalescence by considering viscosity ratio and interfacial tension. Wu [39] has shown that the dispersed drop size is directly proportional to the interfacial tension and to the ±0.84 power of viscosity ratio. For incompatible systems like PP/PE, the interface is too weak due to high interfacial tension and can be easily broken. Therefore by controlling the viscosity of polymers we can decrease the coalescence. Even though small spheres have large surface area and hence more stable, coalescence (which is a non-spontaneous process) occurs in almost all incompatible systems. Thus there must be a driving force for this process. This may be the genuine attractive force between the like particles and/or the interfacial mobility of the dispersed phase in terms of statistical theory of polymer blends.

In H_{20} blend (Fig. 2b) where the concentration of HDPE is 20 wt.%, HDPE domains are much larger than those in H_{10}. As explained earlier, this is partially due to the coalescence phenomenon. Further addition of HDPE results in larger domains as seen in the blends containing 30% HDPE (Fig. 2c) A co-continuous morphology starts when the HDPE is 40%, and for 50/50 blend there is a true co-continuous morphology as shown in Fig. 2d. Up to 40/60 blend this stage persists.

A phase inversion is observed for the 30/70 blends (Fig. 2e) (now HDPE is the major phase). It is interesting to note that the size of domains is smaller when PP is the major phase. This observation can be explained in terms of viscosity difference between the two polymer phases. Being more viscous, PP matrix hinders the diffusion of HDPE domains, so it is difficult for HDPE domains to diffuse through the PP matrix and this limits the number of effective collisions. Hence the phenomenon of coalescence becomes minimum when PP is the matrix. On the other hand it is comparatively easy for the PP domains to diffuse through the less viscous HDPE matrix. So the rate of collisions and therefore the coalescence is greater when HDPE is the matrix.

3.2. Crystallisation and melting behaviour of PP/HDPE blends

The melting and crystallisation behaviours of PP/HDPE blends were analysed calorimetrically. The DSC heating curves of H_0, H_{20}, H_{30}, H_{50}, H_{80} and H_{100} are given in Fig. 4 and the cooling curves are shown in Fig. 5. Melting endotherms and crystallisation exotherms were used to characterise the following parameters:

1. the crystalline melting point (T_m) corresponding to first heating and second heating;
2. the onset temperature of melting;
3. the onset temperature of crystallisation (T_{onset});
4. the crystallisation temperature (T_c) corresponding to first cooling and second cooling;
5. the enthalpy of crystallisation (ΔH_c), determined from the area under the exotherm;
6. heat of fusion (ΔH_f) measured from the area under the endotherm;
7. the percentage crystallinity of HDPE and iPP in blends, compared to 100% crystalline HDPE and iPP, calculated using the relationship:

   $$\%\ \text{Crystallinity} = (\Delta H_f^{obs}/\Delta H_f^0)100$$

   where ΔH_f^{obs} is the observed heat of fusion values and ΔH_f^0 is the heat of fusion value for 100% crystalline HDPE (287 J/g) or iPP (207 J/g).

The results are depicted in Table 1. From the table it is seen that the crystallisation melting point (T_m) of neat PP and HDPE are 165 and 133 °C respectively. The blends show two melting endotherms corresponding to PP and HDPE phases. The crystalline melting point is almost same in all blends during first heating and second heating. This reveals that the blending has no effect on the melting points of PP and HDPE. This also shows that the two polymers are highly immiscible and the blends are incompatible. The onset temperatures of melting and crystallisation of individual polymers are not appreciably affected by blending. However compared to first heating, the melting temperature is slightly increased on second heating. The crystallisation temperature (T_c) of neat PP and HDPE for the first cooling are 109 and 105 °C respectively and for the second cooling the values are 115 and 113 °C respectively. Interestingly, the blends show only one crystallisation temperature (T_c) which are in most case intermediate to those of PP and HDPE.

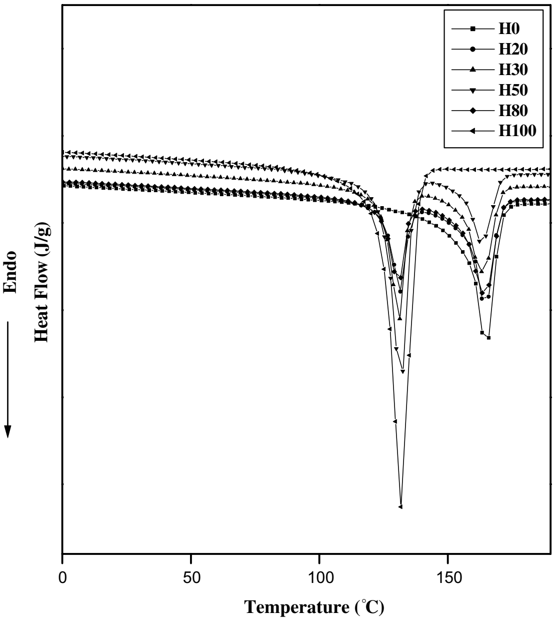

Fig. 4. DSC heating curves showing the crystallisation melting points of H_0, H_{20}, H_{30}, H_{50}, H_{80} and H_{100}.

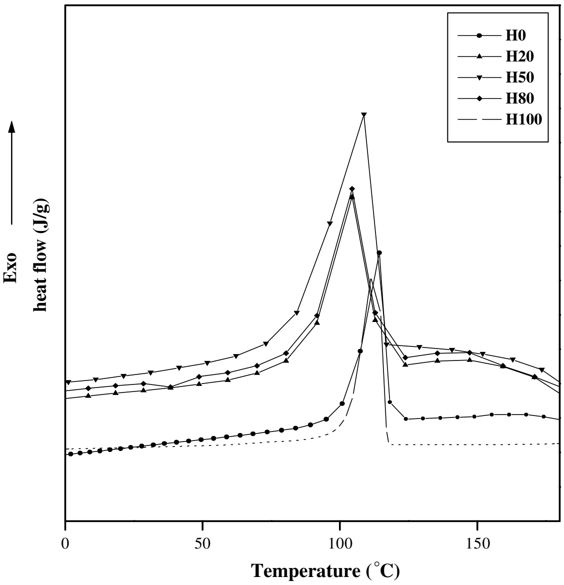

Fig. 5. DSC cooling curves showing the crystallisation temperature of H_0, H_{20}, H_{50}, H_{80} and H_{100}.

From Table 1 it is seen that the addition of PP into HDPE decreases the total enthalpy of crystallisation. Thus maximum energy is liberated during the crystallisation of neat HDPE. This is expected because in virgin HDPE, the nucleation and growth of individual crystallites are so rapid that a very large number of crystallites are formed together so that a large amount of heat is liberated. On the other hand in PP, the nucleation and growth rates are comparatively slow that the individual crystallites get enough time to grow into large spherulites. So large spherulites are formed in PP and this limits the total number of crystallites in a given sample during crystallisation. So the energy liberated during the crystallisation of PP is less compared to that of HDPE. Keith and Padden [40] have shown that in PE there is a very high density of intercrystalline links, but in PP there are only a few such links.

The variation of percentage crystallinity of HDPE and PP in the blend is compared with that of the corresponding theoretical values (calculated from the

Table 1
Melting and crystallisation behaviour of HDPE/PP blends

Blend	Crystalline melting point of HDPE and PP (°C)		Onset temperature (°C) of HDPE and PP		Crystallisation temperature (°C) of HDPE and PP		Enthalpy of crystallisation ($-\Delta H_c$ J/g)	Heat of fusion (ΔH_f J/g)		Percent crystallinity (%)	
	First heating	Second heating	Melting	Crystallisation	First cooling	Second cooling		HDPE	PP	HDPE	PP
H_{100}	132.5	133.0	117.4	120.3	106.5	112.0	165.7	180.0	0	62.7	0
H_{90}	132.5	133.0	118.2	124.0	107.5	115.5	158.7	168.8	44.0	58.8	21.3
	163.0	164.0	147.5								
H_{80}	133.0	133.0	117.3	121.4	106.5	115.5	134.9	174.0	46.0	60.6	22.2
	163.0	164.0	147.2								
H_{70}	133.0	133.0	117.0	121.5	108.0	116.0	119.5	163.6	41.3	57.0	20.0
	163.0	163.0	147.5								
H_{60}	131.5	132.0	116.5	120.5	105.5	113.5	104.4	140.1	43.5	48.8	21.0
	162.5	163.0	145.7								
H_{50}	132.5	133.0	115.5	121.0	105.0	116.0	93.5	124.4	61.2	43.3	29.6
	164.0	165.0	145.4								
H_{40}	132.5	132.0	117.0	121.5	106.5	116.0	84.5	103.8	74.2	36.2	35.8
	163.5	163.0	146.0								
H_{30}	131.5	133.0	116.5	120.5	108.0	115.5	83.2	105.5	78.1	36.8	37.7
	162.5	164.0	145.7								
H_{20}	131.5	133.0	118.2	121.4	105.5	116.0	81.0	101.0	81.4	35.2	39.6
	162.5	163.0	147.5								
H_{10}	132.5	132.0	117.0	121.0	106.5	115.5	83.0	102.0	77.8	35.5	39.5
	163.0	163.0	147.5								
H_0	164.0	165.0	146.5	123.0	112.5	120.0	88.5	0	92.0	0	44.4

weight fraction) in Fig. 6. The percentage crystallinity of HDPE and PP used for the present study are found to be 62.7% and 44.4%. From the figure it is clear that addition of PP decreases the crystallinity of HDPE and vice versa. Thus as the amount of one crystalline polymer in the blend increases, the crystallinity of the second polymer decreases. However it is important to note that the experimental values are slightly smaller than the calculated values.

Based on the variation of crystallinity of HDPE upon addition of PP, we can roughly classify the blends into three groups viz. (a) H_{100} to H_{70} (b) H_{60} to H_{40} and (c) H_{30} to H_0. In the first type, the reduction in crystallinity of HDPE upon addition of PP is not appreciable (∼6%). In the second case, there is abrupt decrease in the crystallinity of HDPE (∼20%) and in the final case, the crystallinity of HDPE remains almost unchanged. Similar trend is observed for the crystallinity of PP. There is no appreciable decrease in the crystallinity of PP up on the addition of 40 wt.% of HDPE (∼6%). However, crystallinity reduces substantially up to H_{70} (∼16%) and further addition of HDPE does not change the crystallinity of PP. These results clearly provide a direct relationship between the crystallinity of a polymer in the blend and the type of morphology of the blend. In the present case, when the polymer forms the continuous phase, its crystallinity is not affected considerably by the presence of the second polymer. Similarly, when one polymer forms the dispersed phase, its crystallinity in the blend is not appreciably affected by the addition of the other polymer. However, when two polymers form a co-continuous phase structure, crystallinity is considerably affected by the presence of the second polymer. Thus the type of morphology decides the crystallisation behaviour of the blend to a certain extent. So we can predict that for polymer blends containing semi-crystalline polymers, final properties are functions of morphology as well as crystallinity and these are intimately related.

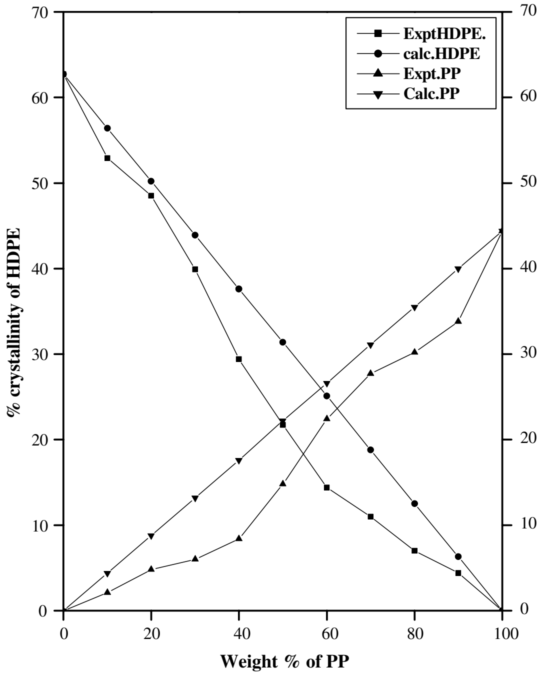

Fig. 6. Variation of percentage crystallinity of HDPE and PP in the blend.

3.3. Mechanical properties of PP/HDPE of blends

The mechanical properties of the blends are presented in Table 2. It is evident from the table that all the properties except hardness show negative deviation. This is due to the incompatibility of the blends. On the other hand, Fig. 7, which gives the variation of Young's modulus with composition, shows a positive synergism with maximum corresponding to 80% PP. This behaviour also can be explained by considering both the morphology and crystallinity of the blends.

For incompatible blends containing at least one semi-crystalline component, the final tensile properties are determined by two competing factors; one is the increase in crystallinity due to the presence of more crystalline component and the other is the extent of compatibility between the two component polymers. The former is the property determining factor at low strain level and the second factor, i.e., the incompatibility, determines the final tensile properties at high strain level. Thus the Young's modulus, which is measured at low strain, shows a positive synergism because here compatibility does not play a significant role. Addition of HDPE decreases the spherulite size of PP by occupying the interspherulite region and thereby decreases the chance for fracture to occur. But the blends are not compatible due to poor interfacial adhesion between the component polymers. Hence stress transfer does not take place between the components. So at high strain, property deterioration takes place as incompatibility leads cracks and fracture at interphase boundaries.

The notched Izod impact strength of the blends is obtained from Table 2. The impact toughness is often the deciding factor in material selection because impact test measures the ability of a polymer to withstand the load imposed upon being struck by an object at high velocity. Thus it is a measure of the energy required to propagate a crack across the specimen. Therefore, the impact properties of these blends are especially important. Addition of higher amount HDPE increases the impact strength of polypropylene. This is not unexpected because HDPE has much lower value of glass transition temperature, ∼−110 °C [41] than that of iPP, ∼10 °C [42]. But due to the incompatible nature of the blend, the increase is actually seen in HDPE rich blends.

Table 2
Mechanical properties of PP, HDPE and their blends

Blend	Tensile strength (MPa)	Tensile strength at break (MPa)	Elongation at break (%)	Impact strength (KJ/m^2)	Hardness (Shore D)	Tear strength (N/mm)
H_0	36.0	30.7	15	21.9	78	7.3
H_{10}	30.3	27.7	12	13.8	77	6.5
H_{20}	28.0	27.5	10	11.4	76	5.0
H_{30}	24.5	24.1	10	8.3	75	4.3
H_{40}	19.6	19.3	10	7.6	73	3.0
H_{50}	19.0	18.6	10	14.8	71	2.9
H_{60}	18.0	15.2	9	15.5	70	2.9
H_{70}	22.8	21.7	9	25.3	69	2.5
H_{80}	23.5	22.5	11	48.8	68	1.6
H_{90}	26.8	26.7	12	53.3	67	2.9
H_{100}	28.7	28.6	20	55.6	63	3.2

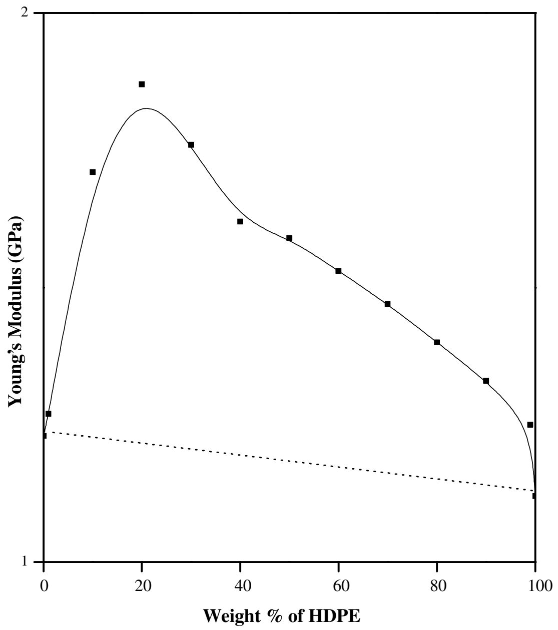

Fig. 7. Effect of addition of HDPE on the Young's modulus of the blends.

This is because on applying a load, cracks originate earlier than expected due to the incompatible nature of the blends. But for blends such as H_{90}, and H_{80} where HDPE is the matrix, load applied is experienced on the HDPE matrix. The increase in the impact properties of PP on the addition of HDPE is related to the crystallinity changes arising due to blending. The addition of HDPE decreases the size of PP spherulites by acting as a nucleating agent. This behaviour can also be explained based on the fact that when HDPE enters the interspherulite boundaries of PP, the HDPE grows very rapidly and deforms the interface between the two components and thus increases the interfacial thickness and enhances the property.

It is important to determine the resistance of the polymer blends to surface damage or to plastic deformation. Thus hardness measurements are necessary. Table 2 gives the hardness of HDPE, PP and their blends. The hardness values range from 63 to 78 Shore D. Neat HDPE shows the minimum Shore D hardness and pure PP shows the maximum. On adding HDPE into PP, the hardness decreases as expected. The hardness values lie above the additivity line. This is because being a surface property, hardness is less related to the compatibility of the blends.

The tear test is relatively a simple method for evaluating the tearing behaviour of polyolefins. Fig. 8 gives the load-displacement curves of PP, HDPE and their blends. The tear strength values of PP, HDPE and their blends are depicted in Table 2. The maximum tear strength is for neat PP. This shows that virgin PP offers maximum tearing forces. Hence PP tears at highest force and at the smallest displacement. On adding HDPE to PP the tear strength decreases because HDPE increases the ductility of the blends. As the amount of HDPE increases the tear strength decreases and reaches a minimum at 80 wt.% of HDPE. Neat HDPE has more tear strength than the blends in which HDPE is the matrix (H_{70}, H_{80} and H_{90}), even though we expect the opposite due to the more ductile nature of the virgin sample. This is because of two effects. The first one is the incompatibility of the blends and the second is the low tear strength of HDPE compared to PP. In neat PP these two effects are absent. In PP rich blends since PP is the matrix phase we can neglect the second factor; thus the decrease of tear strength is mainly due to the incompatibility. But for HDPE rich blends both these factors simultaneously operate and the effect is maximum for H_{80}.

All these studies reveal that the mechanical properties of polymer blends containing at least one

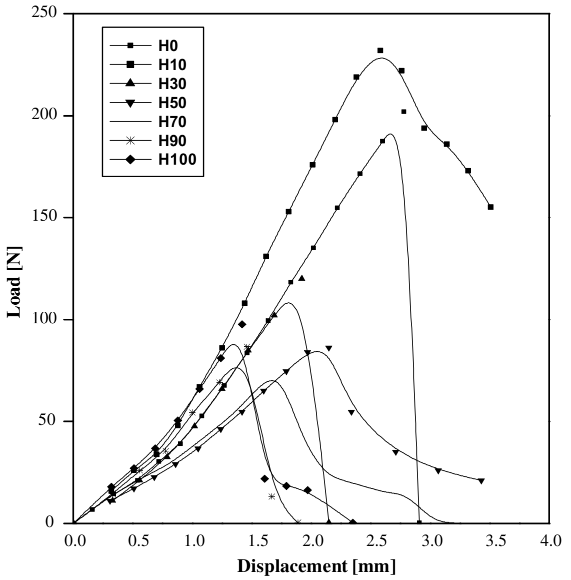

Fig. 8. Load vs. displacement of PP, HDPE and their blends.

semi-crystalline polymer strongly depend on the morphology and crystallinity changes that occur during blending. The immiscibility and incompatibility of the blends may be explained in terms of morphological features and crystallisation behaviour derived from the present study. As explained earlier, most of the mechanical properties show negative deviation due to the immiscibility and incompatibility of the blends. But the 80/20 PP/HDPE blend shows synergism in Young's modulus. It was reported that during crystallisation of the matrix, deformation of the dispersed particles occurs and this results in the deformation of the interface in polymer blends. Thus during the crystallisation of the blend where PP is the matrix phase, solidification of PP occurs in presence of HDPE melt which constitutes the dispersed phase. During this process, molten HDPE flows into the region between iPP spherulites growing near the interface. This results in the deformation of the interface between PP and HDPE, and the deformation ends with the completion of crystallisation of spherulites, when all the polypropylene melt is converted to spherulites. The net result is an increase in interfacial area. Thus more stress transfer occurs in the case of 80/20 blend. But unfortunately the formed interfaces are so weak that they can transfer stresses only at very low strain levels. This is why this blend shows synergism in Young's modulus, which is measured, at low strain levels.

In the case of 90/10 blend as evidenced from the scanning electron micrographs, the HDPE concentration is too low to create deformation at the interfaces. On the other hand for 70/30 blend another phenomenon called, the clustering of individual polymers, is more pronounced. It was established that at temperatures above the crystallisation melting points of homopolymers, PE and PP chains are found to segregate into distinct domains and the PE phase shows more significant clustering [43]. The clustering of PE and PP is due to the unfavourable cross-correlations between PE and PP chains in the blend [44]. This unfavourable clustering increases as the amount of individual polymers increases in the blend. Thus the immiscibility may be maximum for blends containing 40–60 wt.% of HDPE. This may be the plausible explanation for the minimum mechanical properties observed for blend containing 40–60 wt.% of HDPE. Thus from these studies it is clear that each and every changes in the mechanical properties of the blends are the results of changes in their phase morphologies which in turn is a factor of crystallinity if at least one component is semi-crystalline.

4. Conclusions

From the present study, which is devoted to determine the phase morphology, crystallisation behaviour and mechanical properties of iPP/HDPE blends, the following conclusions can be drawn:

1. The phase morphology obtained from the scanning electron micrographs of fracture surfaces of the blends revealed a non-uniform and unstable morphology owing to the highly incompatible nature of the homopolymers.
2. Melting and crystallisation behaviour of the blends suggested that blending has no considerable effect on the melting point and crystallisation temperature of the individual polymers. When HDPE or PP forms the dispersed phase or matrix, the crystallinity of the polymer is not affected considerably by the second polymer, while when these form a co-continuous phase structure, the crystallinity of one polymer is strongly affected by the second one.
3. Mechanical properties of the blends showed that all the properties except Young's modulus and hardness show negative deviation from additivity line and minimum values of mechanical properties are observed for blends exhibiting co-continuous morphology.
4. Finally it can be concluded that the mechanical properties of the blends are intimately dependent on each and every change in the phase morphology and crystallisation behaviour.

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